

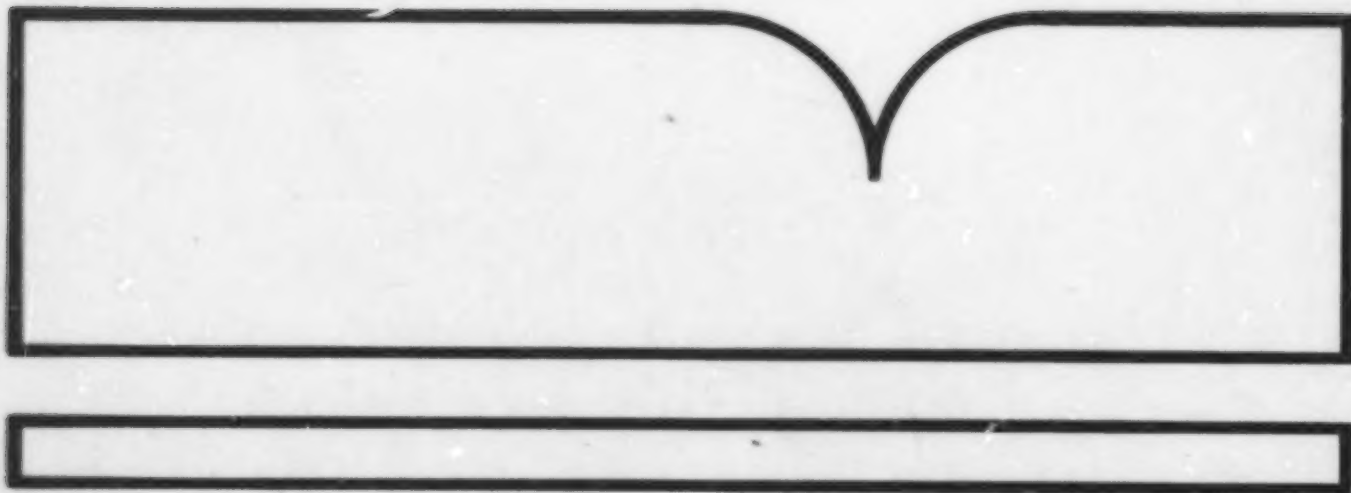
Observations of the Modification of Cloud  
Condensation Nuclei in Wave Clouds

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## OBSERVATIONS OF THE MODIFICATION OF CLOUD CONDENSATION NUCLEI IN WAVE CLOUDS<sup>(\*)</sup>

by

Dean A. HEGG, Peter V. HOBBS, Lawrence F. RADKE  
Atmospheric Sciences Department (\*\*)  
University of Washington  
Seattle, Washington 98195, U.S.A.

### RÉSUMÉ

*On présente des mesures qui montrent, sous le vent de nuages d'onde, des concentrations accrues de noyaux de condensation nuageuse, actifs pour une gamme de sursaturations. Des mesures simultanées des spectres granulométriques de particules corroborent ces mesures et suggèrent qu'il y a augmentation de la dimension des particules à l'intérieur des nuages. Les modifications dans les spectres sont plus prononcées vers un diamètre de 1  $\mu\text{m}$ . Les résultats obtenus suggèrent que ces observations peuvent s'expliquer par la production de sulfate dans les nuages.*

### ABSTRACT

*Measurements are presented which show enhanced concentrations of cloud condensation nuclei, active at various supersaturations, downwind of wave clouds. Concurrent measurements of particle size spectra corroborate these measurements and suggest particle growth within the clouds. Modifications in the particle size spectra are most pronounced at diameters of  $\sim 1 \mu\text{m}$ . Measurements and modeling results suggest these observations can be explained by sulfate production in clouds.*

### 1. — INTRODUCTION.

There is a growing body of evidence that the particles released by evaporating clouds differ from those that act as cloud condensation nuclei (CCN) during the formation of the clouds. Thus, Radke and Hobbs (1969), Saxena *et al.* (1970), Dinger *et al.* (1970) and Radke (1970) observed that the concentration of CCN active at a given supersaturation are often higher in air from evaporating clouds than in the ambient air.

(\*) This article represents a portion of the paper entitled « A Preliminary Study of Cloud Chemistry » presented at the VIII<sup>e</sup> Conférence Internationale sur la Physique des Nuages, Clermont-Ferrand, France, 15-19 July, 1980, which was printed on pages 7-10 of the Preprint Volume.

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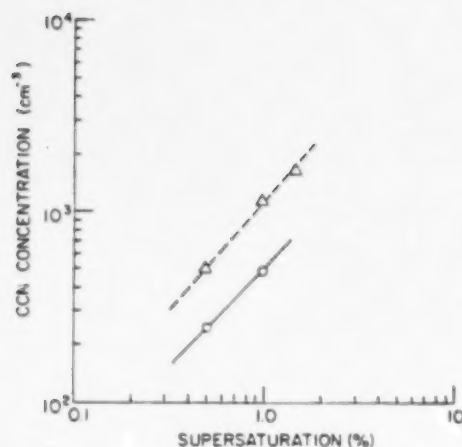


FIG. 1. — Cloud condensation nucleus (CCN) downwind (dashed line) and upwind (solid line) of a wave cloud on May 3, 1979.

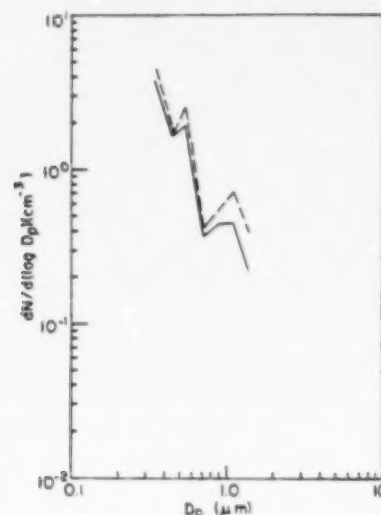


FIG. 2. — Particle size spectra measured downwind (dashed line) and upwind (solid line) of a wave cloud on May 3, 1979.

Two possible explanations for these modifications have been proposed. Firstly, if chemical reactions take place within cloud droplets the products of these reactions should be deposited onto the original CCN when the droplets evaporate (Radke and Hobbs, 1969; Easter and Hobbs, 1974; Hegg and Hobbs, 1979). Alternatively, CCN may shatter during the final phases of droplet evaporation, as suggested by the early laboratory work of Dessens (1946). If the first mechanism is operative, there should be an increase in the concentration of CCN at all supersaturations after processing by a cloud. On the other hand, if shattering were responsible, the concentrations of CCN above a certain supersaturation should increase but at lower supersaturations they should decrease. Also, in the case of the former mechanism (but not the latter), the number concentration of particles from a few tenths up to a few micrometers in diameter should increase (due to the more numerous small particles moving up in size as the result of the deposition of soluble material) and the total particle mass (or volume) should be greater in the air leaving the cloud than in the air entering the cloud. By contrast, Radke and Hegg (1972) found in laboratory experiments that particles greater than just a few tenths of a micrometer in size decreased in number due to shattering.

The data obtained in the field studies referred to above are not adequate to distinguish between these two mechanisms for CCN modification because the measurements were restricted to CCN concentrations at one supersaturation. In an effort to obtain more definitive information on the processes involved, we have carried out a new series of field measurements in which both CCN and particle size spectra have been obtained in air entering and leaving wave clouds.

## 2. — EXPERIMENTAL.

All measurements reported here were obtained aboard the University of Washington's B-23 research aircraft. The particle-measuring instrumentation and data processing techniques have been described in detail by Hobbs *et al.* (1976, 1979). The

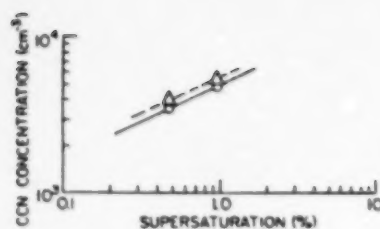


FIG. 3. — Cloud condensation nucleus spectra measured downwind (dashed line) and upwind (solid line) of a wave cloud on May 10, 1979.

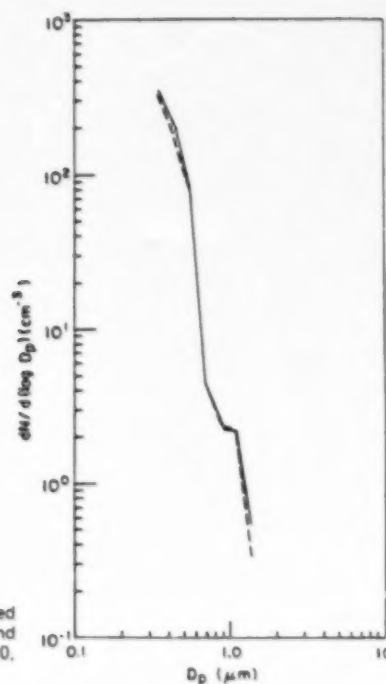


FIG. 4. — Particle size spectra measured downwind (dashed line) and upwind (solid line) of a wave cloud on May 10, 1979.

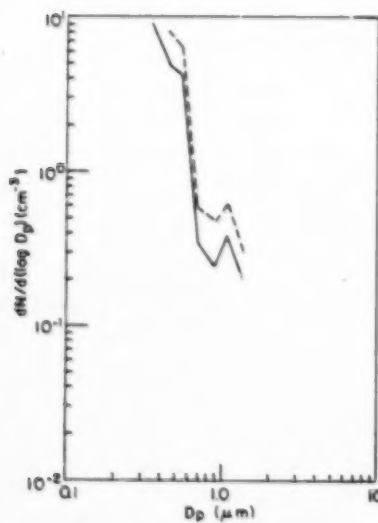


FIG. 5. — Particle size spectra measured downwind (dashed line) and upwind (solid line) of a wave cloud over Mt Rainier on July 13, 1979.



CCN concentrations are measured as a function of supersaturation in a thermal gradient diffusion chamber. Particle size distributions are determined by measurements with a diffusion battery - Aitken nucleus counter combination, and two Royco optical particle counters. The optical counters are calibrated on the assumption that the refractive index of the particle is 1.5, which is similar to the value for many atmospheric aerosols. While spatial inhomogeneities in the index of refraction may produce small, spurious, spectral changes in such instruments, the spatial inhomogeneity at our sampling height of  $\sim 3$  km is too small to produce any effects of consequence. It should be noted that the particles sampled by the B-23 measuring system are put through a diffusion dryer prior to analysis to ensure that the measured particle sizes are those of the *dry* particles.

The airborne procedure was to first measure CCN and particle size spectra in the clear air just downwind of an evaporating trailing edge of a cloud and then to repeat these measurements in clear air just upwind of the condensing, leading edge of the cloud. Finally, cloud droplet size distributions and liquid water contents were measured within the cloud.

### 3. — RESULTS AND DISCUSSION.

We note first that we have obtained evidence for CCN enhancement in only about one-third of the 17 wave clouds we have studied. Lack of a detectable CCN enhancement in the remaining cases may have been due to sampling problems (e.g. incorrect identification of the inflow and/or outflow regions of the cloud) or to the lack of any appreciable CCN enhancement in these particular clouds. For example, it is well substantiated that several of the possible sulfate production mechanisms in the aqueous phase depend critically on the presence of various trace components in the air (Hegg and Hobbs, 1978). The wave clouds we studied were relatively high ( $\sim 3$  km) and situated in fairly clean air over Mt. Rainier, Washington, where these trace components may not have always been present.

An example of CCN spectral enhancement by a wave cloud is shown in Figure 1. The simultaneous enhancement of the CCN concentrations at several supersaturations suggests that deposition of soluble material on the CCN involved in the formation of the cloud was responsible for the enhanced CCN activity downwind. Particle size spectra measurement upwind and downwind of this cloud (Fig. 2) support this conclusion since there is a general enhancement in the number concentrations of particles over a size range from  $\sim 0.3$  to  $2 \mu\text{m}$ .



It can be seen from Figure 2 that the increase in particle concentrations was most pronounced at a particle diameter of  $\sim 1 \mu\text{m}$ . This  $1 \mu\text{m}$  « peak » is typical of cases in which we have observed enhanced CCN concentrations downwind of wave clouds. Indeed, the size of the  $1 \mu\text{m}$  peak seems to be related to the degree of CCN enhancement. For example, Figure 3 shows a case of very little CCN enhancement, and the particle size spectrum associated with this case (Fig. 4) shows no discernable peak at  $\sim 1 \mu\text{m}$ . However, care must be exercised in linking the  $1 \mu\text{m}$  peak directly to the CCN enhancement we have observed. The largest CCN we have measured, those active at 0.5 % supersaturation, should have a dry diameter of only  $\sim 0.1 \mu\text{m}$ . Therefore, it appears that the particles producing the  $1 \mu\text{m}$  peak are not directly responsible for the observed enhancement in CCN concentrations. Instead, both phenomena are manifestations of the production of particulate mass in cloud droplets and of its deposition onto CCN when the clouds evaporate.

A case study of a wave cloud over Mt. Rainier on July 13, 1979, provides further information on the nature of the production process. The  $\sim 1 \mu\text{m}$  peak characteristic of mass production within clouds is well defined in this case (Fig. 5), although no corroborating CCN measurements were made on this flight. Filter samples revealed that the concentration of particulate sulfate in the air was  $0.68 \mu\text{g}/\text{m}^3$  higher downwind than it

was upwind of this wave cloud. The particle enhancement displayed in Figure 5 (and the presumed CCN enhancement) can therefore be interpreted in terms of  $\text{SO}_2$ -to- $\text{SO}_4^*$  conversion, as described theoretically by Easter and Hobbs (1974) and Hegg and Hobbs (1979). The Easter-Hobbs interactive cloud-chemistry model (as modified by Hegg and Hobbs), run for initial conditions similar to those for July 13, 1979, revealed a change in particulate sulfate due to cloud processing of  $0.5 \mu\text{g}/\text{m}^3$ . The CCN enhancement predicted by the model for this case is shown in Figure 6. The change in CCN spectrum displayed here is similar, in terms of percentage increase in CCN active at given supersaturations, to those we have observed in wave clouds.

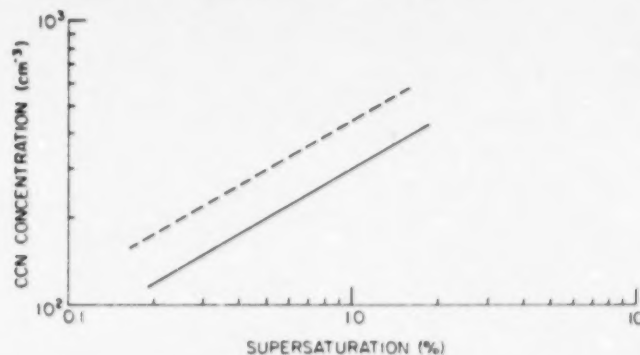


FIG. 6. — Cloud condensation nucleus (CCN) spectra downwind (dashed line) and upwind (solid line) of a wave cloud as predicted by the numerical model of Easter and Hobbs (1974) for conditions similar to those observed in the field on July 13, 1979.

The case of July 13, 1979 is the only one for which we have sufficient chemical data to run the Easter-Hobbs models and also good measurements of particle spectra. However, we have several additional data sets with sufficient chemical measurements for the model predictions of sulfate production to be compared with the observations. These latter results, which support the conclusion that sulfate is produced in wave clouds, are presented elsewhere (Hegg and Hobbs, 1981).

#### 4. — CONCLUSIONS.

Measurements upwind and downwind of wave clouds show that in about one-third of the cases the concentrations of cloud condensation nuclei, particles from a few tenths of a micrometer to  $2 \mu\text{m}$  in diameter, and the mass of sulfate in the air, are greater downwind of the clouds than upwind. These measured enhancements are consistent with a particle mass production mechanism within the cloud involving  $\text{SO}_2$ -to- $\text{SO}_4^*$  conversion in the cloud droplets. Moreover, the CCN spectral enhancement and sulfate production are in good agreement with the predictions of the interactive cloud-chemistry model for this process developed by Easter and Hobbs (1974).

#### ACKNOWLEDGMENTS.

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